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PATENT

Attorney Docket No. 29610/CDT091

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: Berger *et al.* Serial No. 10/009,078 Filed: June 1, 2000 For: Method of Producing Organic Light-Emissive Devices Group Art Unit: To be assigned Examiner: To be assigned	I hereby certify that this paper is being deposited with the United States Postal Service as first class mail, postage prepaid, in an envelope addressed to: Assistant Commissioner for Patents, Washington, DC 20231 on this date: March 28, 2002 James P. Zeller, Reg. No. 28,491

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Assistant Commissioner for Patents
Washington, DC 20231

Sir:

Submitted herewith are certified copies of Great Britain application No. 9913451.2 filed June 9, 1999 and Great Britain application No. 9913695.4 filed June 11, 1999, the priority of which are claimed under 35 U.S.C. § 119.

Respectfully submitted,

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March 28, 2002

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2.	Patent application number *(The Patent Office will fill in this part)*	**9913451.2** 09 JUN 1999
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	Patents ADP number *(if you know it)*	6166441002
	If the applicant is a corporate body, give the country/state of its incorporation	**UNITED KINGDOM**
4.	Title of the invention	**LIGHT-EMITTING DEVICES**
5.	Name of your agent *(if you have one)* "Address for service" in the United Kingdom to which all correspondence should be sent *(including the postcode)*	**PAGE WHITE & FARRER** **54 DOUGHTY STREET** **LONDON** **WC1N 2LS** **UNITED KINGDOM**
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		Country	Priority application number *(if you know it)*	Date of filing *(day / month / year)*
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91352

LIGHT-EMITTING DEVICES

This invention relates to light-emitting devices, for example devices suitable as display devices.

One specific class of display devices is those that use an organic material for light emission. Light-emissive organic materials are described in PCT/WO90/13148 and US 4,539,507, the contents of both of which are incorporated herein by reference. The basic structure of these devices is a light-emissive organic layer, for instance a film of a poly(p-phenylenevinylene ("PPV"), sandwiched between two electrodes. One of the electrodes (the cathode) injects negative charge carriers (electrons) and the other electrode (the anode) injects positive charge carriers (holes). The electrons and holes combine in the organic layer generating photons. In PCT/WO90/13148 the organic light-emissive material is a polymer. In US 4,539,507 the organic light-emissive material is of the class known as small molecule materials, such as (8-hydroxyquinoline)aluminium ("Alq3"). In a practical device one of the electrodes is typically transparent, to allow the photons to escape the device.

Figure 1 shows the typical cross-sectional structure of an organic light-emissive device ("OLED"). The OLED is typically fabricated on a glass or plastic substrate 11 coated with a transparent first electrode 12 such as indium-tin-oxide ("ITO"). Such coated substrates are commercially available. This ITO-coated substrate is covered with at least a layer of a thin film of an electroluminescent organic material 13 and a final layer forming a second electrode 14, which is typically a metal or alloy. Other layers can be added to the device, for example to improve charge transport between the electrodes and the electroluminescent material.

One advantage of typical OLEDs is that the field of light output from the device is generally Lambertian - i.e. from each part of the device in which emission occurs photons are emitted uniformly in all directions. This is a highly advantageous

feature for display devices for general use because it gives rise to an image that is uniform from a wide range of viewing angles.

The emission from OLEDs can be enhanced by arranging for the emitting region of the device to be located in a resonant cavity. The resulting structure is a planar cavity acting as a Fabry-Perot resonator with two mirrors spaced apart by a cavity which contains a photon-emitting material 4. The mirror separation is set to be of the order of the optical wavelength, so that the resonant frequency of the cavity corresponds to an optical frequency. Such a structure therefore has a narrow emission spectrum, allowing emission only at the resonance wavelength(s) of the cavity. It is also capable of enhancing the emission at a certain wavelength compared to the free-space emission of the luminescent material (see J. Grüner et al. J Appl. Phys. 80, 207 (1996)). It would be desirable for display devices including OLEDs to make use of these advantages of resonant cavities. However, the light output from such a resonant cavity is generally unidirectional and not Lambertian. Therefore, the advantages of the resonant cavity are to a large extent outweighed by the deterioration in viewing angle and image uniformity.

According to one aspect of the present invention there is provided a light-emitting device comprising: a pair of electrodes; a light-emitting region located between the electrodes and comprising light-emitting organic material; a first reflective layer and a second reflective layer, the second reflective layer being partially light-transmissive and the first and second reflective layers being located on either side of the light-emitting region to define a resonant cavity structure about the light-emitting region; and a light-dispersal structure outside the cavity for dispersing light emitted from the cavity through the second reflective layer.

According to a second aspect of the present invention there is provided a method for forming a light-emitting device having a pair of electrodes; a light-emitting region located between the electrodes and comprising light-emitting organic material; and a first reflective layer and a second reflective layer, the second reflective layer being partially light-transmissive and the first and second reflective

layers being located on either side of the light-emitting region to define a resonant cavity structure about the light-emitting region; the method comprising forming a light-dispersal structure over the second reflective layer for dispersing light emitted from the cavity through the second reflective layer.

The light-dispersal structure is suitably capable of refracting light emitted from the cavity. Thus the light-dispersal structure preferably includes scattering features of a scale in the range from 0.1 to 0.5 times the resonance wavelength of the cavity. The light-dispersal structure is suitably capable of scattering light emitted from the cavity. The light-dispersal structure is suitably capable of increasing the viewing and/or emission angle of the display, preferably to 90° or more.

The light-dispersal structure suitably comprises discrete regions of two or more materials having different refractive indices. One of the materials may take the form of a matrix and is preferably light-transmissive or transparent. Another material may be dispersed in discrete regions in the matrix - for instance as particles or voids. The particles or voids could be of any shape, for example generally spherical. The said step of forming the light-dispersal structure suitably comprises depositing or otherwise forming a light-transmissive material including a dispersal of light-transmissive particles. The dispersed material is suitably light-transmissive – i.e. fully or partially transparent. The regions of the dispersed material may be fully or partially reflective.

The light-dispersal structure is suitably in the form of a layer. Such a layer suitably has a major surface that is uneven, whereby diffusion by refraction of light from the cavity may occur at the interface of that surface with air or with an adjacent layer of the device. The said major surface is suitably the major surface further from the cavity. The unevenness of the surface may take the form of surface roughness, grooves and/or ridges.

The device may have a coating of an encapsulant or protective layer. The encapsulant or protective layer is suitably located between the second reflective

layer and the light-dispersal structure. Alternatively the encapsulant or protective layer could be located between the second reflective layer and the light-dispersal structure, or the light dispersal structure could be formed in and/or integrated with the encapsulant or protective layer.

Either or both of the first and second reflective layers may be formed as a distributed Bragg reflector (which could be electrically conductive or electrically insulating) or by a respective one of the electrodes or in another way (e.g. by additional metallisation).

Either or both of the electrodes may be in the form of a layer. Preferably the layers of the electrodes are parallel with those of the reflectors. One or both of the reflectors may be located between the electrodes. One or both of the electrodes may be located between the reflectors.

One of the electrodes may be an anode electrode and the other a cathode electrode. Either or both of the electrodes is preferably at least partially light-transmissive, most preferably substantially transparent, at least to light of some or all of the wavelengths that can be emitted from the device.

One of the electrodes suitably has a work function greater than 4.0eV. The electrode could, for example, be formed of ITO (indium-tin oxide), TO (tin oxide) or gold. The electrode could be patterned to allow individual regions (e.g. pixels) of the display to be individually addressed.

One of the electrodes suitably has a work function below 3.5eV. The electrode could, for example, be formed of a low work-function metal such as Li, Ca, Mg, Cs, Ba, Yb, Sm etc. Ca, or a fluoride or oxides of such a metal together, optionally, with a conductive material such as Al, which could be co-deposited or sequentially deposited with the metal, oxide or fluoride. Where the electrode is at least partially light-transmissive this could be achieved by forming the electrode from a light-

transmissive material and/or by the electrode being relatively thin, for example less than 2, 5, 10, 20 or 30nm in thickness.

The light-emitting material is preferably a polymer material, most preferably a semiconductive and/or conjugated polymer material. Alternatively the light-emissive material could be of other types, for example sublimed small molecule films. The light emitting material could be or comprise inorganic light-emissive material. The or each organic light-emissive material may comprise one or more individual organic materials, suitably polymers, preferably fully or partially conjugated polymers. Example materials include one or more of the following in any combination: poly(p-phenylenevinylene) ("PPV"), poly(2-methoxy-5(2'-ethyl)hexyloxyphenylenevinylene) ("MEH-PPV"), one or more PPV-derivatives (e.g. di-alkoxy or di-alkyl derivatives), polyfluorenes and/or co-polymers incorporating polyfluorene segments, PPVs and related co-polymers, poly(2,7-(9,9-di-n-octylfluorene)-(1,4-phenylene-((4-secbutylphenyl)imino)-1,4-phenylene)) ("TFB"), poly(2,7-(9,9-di-n-octylfluorene) - (1,4-phenylene-((4-methylphenyl)imino)-1,4-phenylene-((4 - methylphenyl)imino) - 1,4-phenylene)) ("PFM"), poly(2,7 - (9,9 - di-n-octylfluorene) - (1,4-phenylene-((4-methoxyphenyl)imino)-1,4-phenylene-((4-methoxyphenyl)imino)-1,4-phenylene)) ("PFMO"), poly (2,7-(9,9-di-n-octylfluorene) ("F8") or (2,7-(9,9-di-n-octylfluorene)-3,6-Benzothiadiazole) ("F8BT"). Alternative materials include small molecule materials such as Alq3.

One or more charge-transport layers may be provided between the light-emissive region and one or both of the electrodes. The or each charge transport layer may suitably comprise one or more polymers such as polystyrene sulphonic acid doped polyethylene dioxythiophene ("PEDOT-PSS"), poly(2,7-(9,9-di-n-octylfluorene)-(1,4-phenylene-(4-imino(benzoic acid))-1,4-phenylene-(4-imino(benzoic acid))-1,4-phenylene)) ("BFA"), polyaniline and PPV.

Any implied physical orientation of the device is not necessarily related to its physical orientation during use or manufacture.

The present invention will now be described by way of example with reference to the accompanying drawings. In the drawings:

figure 1 shows an organic light-emitting device; and

figure 2 shows an organic light-emitting device including a light-dispersal structure.

The OLED of figure 2 includes a region 1 of light emitting material. An anode electrode 2 and a cathode electrode 3 are located on either side of the light-emitting material. A resonant cavity reflector structure is defined about the light-emitting region 1 by a reflective layer 4 and a partially reflective and partially light-transmissive layer 5. The resonant structure is capable of enhancing emission from the light-emitting region and/or narrowing the emission spectrum from the region. On the outside of the partially light-transmissive layer is a structure 6 that is capable of dispersing the light emitted from the cavity so as to render the far-field emission from the overall device more Lambertian.

The resonant structure of the device constitutes a planar microcavity. The resonance wavelengths, λ_{Res}, of the cavity are determined by the effective length, L_{eff}, of the cavity. The effective length of the cavity is a function of the refractive index and the thickness of each of the layers in the cavity and the phase change on reflection of light from the mirrors. If the refractive index of the kth layer of the cavity is n_k and the thickness of the kth layer of material is d_k then the effective length of the cavity is given by:

$$L_{eff} = L_{phase\_change} + \sum_k n_k \cdot d_k$$

and the resonance wavelengths are given by:

$$\lambda_{Res} = \frac{2L_{eff}}{q}$$

where q is an integer.

The electroluminescent device of figure 2 is formed on a glass substrate 9 which is coated with an anode electrode layer 2 of indium-tin oxide (ITO). Such ITO-coated glass substrates are commercially available. The glass sheet could be a sheet of

sodalime or borosilicate glass of a thickness of, for instance, 1mm. Instead of glass other materials such as Perspex could be used. The thickness of the ITO coating is suitably around 150nm and the ITO suitably has a sheet resistance of between 10 and 30Ω/□, preferably around 15Ω/□. Instead of ITO other materials, preferably of relatively high work function, such as tin oxide (TO) could be used.

A distributed Bragg reflector (DBR) consists of a stack of regularly alternating higher- and lower-refractive index dielectrics (light transmissive materials) fabricated to fulfil the Bragg condition for reflection at particular wavelengths. This occurs when the optical path of the periodicity in the dielectric stack corresponds to half a wavelength, and the reflectivity is further optimised when the DBR stack obeys the following condition:

$$\tfrac{1}{2}\lambda = n_1 d_1 + n_2 d_2,$$

where n_1, n_2 are the respective refractive indices; d_1, d_2 are the corresponding component film thicknesses in the DBR; and λ is the desired reflection wavelength.

The reflective layer 4 is formed over the ITO layer 2. The reflective layer 4 is formed as a distributed Bragg reflector structure formed of alternating layers of conductive or semiconductive materials such as partially doped PPV or other conjugated polymers (see our co-pending UK patent application number 9907802.4, the contents of which are incorporated herein by reference) or GaAs and AlGaAs. The layer thicknesses are chosen so that the structure is capable of reflecting light at a selected emission frequency of the emissive region. Instead of the reflective layer 4 being formed between the electrode 2 and the light-emitting layer 1 the electrode 2 could be formed between the light-emitting layer 1 and the reflective layer. In that case the reflective layer would not have to be electrically conductive, but the electrode 2 should be light-transmissive, as is ITO, for instance. Another alternative is for the reflective structure to be formed over only parts of the ITO layer - in that case the reflector could be electrically insulating but conduction from the ITO to the emissive layer is possible in the regions not covered by the reflective layer. Suitable materials for an electrically insulating

DBR include Si and SiO_2. A further alternative is to form the ITO layer 2 to a thickness of a quarter of the (or a desired) emission wavelength of the light-emitting material, so it acts as at least a partial DBR structure - the separate reflective structure could then be omitted.

In the device of figure 2 a charge transport layer 7 is formed over the reflective layer 4. The charge transport layer improves hole transport between the anode and/or reflective structure 4 and the light-emitting region 1. The charge transport layer is formed of PEDOT:PSS. The charge transport layer could be formed from a solution containing PEDOT:PSS with a ratio of PEDOT to PSS of around 1 to 5. The hole transport layer could be spin-coated from solution and then baked e.g. at 200°C for 1 hour in a nitrogen environment. The thickness of the transport layer is suitably around 50nm but the thickness may be chosen so as to set the width of the resonant cavity to a desired value whilst allowing the thicknesses of the other layers of the device to be optimised for other factors. Instead of PEDOT:PSS other conductive materials such a polyaniline could be used for the charge transport layer 7. Alternatively the charge transport layer could be omitted and the thicknesses of other layers of the device could be chosen so as to fix the width of the cavity.

Then the light emitting layer 1 is deposited. The light-emitting layer could be formed of any suitable organic light-emitting material, but polymer materials are preferred. For example, an electroluminescent layer 12 comprising 20% TFB in 5BTF8 could be coated over the hole transport layer by spin-coating typically to a thickness of 90nm. The term 5BTF8 refers to poly (2,7-(9,9-di-*n*-octylfluorene) ("F8") doped with 5% poly-(2,7-(9,9-di-*n*-octylfluorene)-3,6-benzothiadiazole) ("F8BT").

The cathode electrode layer 3 is formed over the light-emitting layer 1. The cathode electrode layer is formed of LiF with a thickness of 0.5 to 10nm, formed by evaporation of LiF. Such a layer is light-transmissive, so that light can pass through it to leave the device. In the device of figure 2 the reflector 4 is fully

reflective and emission is from the upper surface of the device as illustrated in figure 2. Therefore, the cathode is formed of a light-transmissive material. If the reflector 4 were partially reflective and emission from the lower surface of the device the cathode could be opaque, and formed of e.g. thin layer of Ca covered with a protective layer of Al. It is preferred that the material of the cathode electrode, at least at the surface facing the light-emitting material, has a relatively low work function.

In this embodiment a further DBR structure 5 is formed over the cathode electrode. The structure 5 is formed as for structure 4, but with fewer layers so that the structure 5 is not fully reflective but is partially reflective and partially light transmissive. This allows some leakage from the cavity structure so that light can leave the device. A suitable form for the structure 5 is again alternating layers of differently doped conjugated polymer(s), alternating layers of GaAs/AlGaAs or alternating layers of Si/SiO_2. Since the structure 5 is not located between the electrodes it need not be electrically conductive or semiconductive but could be formed of alternating layers of insulating materials. Alternatively, the cathode layer 3 could be metallised so that it is partially reflective and partially light-transmissive, so that there is no need for a separate reflective layer.

The light dispersal structure 6 is formed over the upper reflector of the cavity - i.e. outside (preferably immediately outside) the cavity in the viewing direction. In the embodiment of figure 2 the light-dispersal structure is formed by coating the device with a layer of light-transmissive polymer material containing a dispersal of light-transmissive particles 8. The particles have a different refractive index from the polymer matrix, so light leaving the device is scattered as it passed through the structure. The particles could be fragments or spheres of glass or latex or nanoparticles of a material such as TiO_2 (see our co-pending UK patent application number 9815270.5, the contents of which are incorporated herein by reference). Suitable materials for the matrix include fluorinated polymers, polyimide, polyethylene or a photoresist material. The diameter of the particles could suitably be 0.1 to 0.5 times the selected emission wavelength of the device.

It may be preferred for the particles to have a size distribution across all or part of that range or outside it in order to lessen any wavelength dependence of the scattering - this may be useful if the device is to be a full-colour display. The thickness of the layer of matrix could be 10-5000nm and the loading of the particles 10-60% by volume. The structure could be formed by spin-coating, spray coating, evaporation or lamination. The materials of the particles and the matrix should be chosen so that they have different refractive indices (so that scattering can occur in the structure) and also so that the refractive index of the matrix does not cause undesirable reflections at its interface with the adjacent underlying layer.

Instead of containing solid particles, the matrix of the light dispersal structure could be formed so as to include bubbles or pockets of air that could similarly perform scattering of the emitted light.

Finally, contacts are made to the anode and cathode electrodes to allow the device to be driven, and the device is encapsulated, for example in epoxy, for environmental protection.

When the device is in use emitted light is scattered by the structure 6, which renders the far-field emission from the device more Lambertian. After scattering, the light is emitted in a range of directions from each point over the device's area. The range of directions is preferably greater than 90° and may preferably approach 180°.

The structure 6 could be formed outside, rather than inside the encapsulant. Forming the structure 6 atop the encapsulant may help to reduce the possibility of waveguiding in the device. If the encapsulant itself provides the scattering structure then it is especially preferable for the scattering to be by means of bubbles or voids (e.g. containing air or inert gas) in the encapsulant layer.

The structure 6 could be formed in many other ways. For example, the structure could be formed by a layer (for example of a polymer material such as fluorinated

polymers, polyimide, polyethylene or a photoresist material) having surface relief on its upper surface (the surface further from the light-emitting region) so that it causes scattering of emitted light there. The surface relief could be formed by etching (e.g. plasma or wet etching) into the surface, either randomly or in a predetermined design, by stamping or scratching or in another way. The predetermined design could be chosen to promote Lambertian output. The upper surface could be uneven or rough, and could include a pattern of scratches, grooves, ridges or other surface irregularity which could be formed by scraping or stamping. The scale of the roughening is suitably in the size range that will cause efficient scattering of the light emitted from the cavity - e.g. in the range from 0.1 to 0.5 times the emission wavelength of the device.

In the embodiment of figure 2 the reflector 5 is partially reflective and the electrode 3 is light transmissive. In other devices the reflector 4 could be partially reflective and the electrode 2 light transmissive.

The electrodes could be reversed, so that the first electrode to be deposited is the cathode and the upper electrode the anode.

A charge transport layer could be provided between the emissive material and the cathode to improve electron transport into the light-emitting region.

Either or both of the electrodes may be patterned so to allow individual regions of the display to be selectively addressed.

The particles could be opaque, provided that they were of a size that nevertheless inherently gave rise to suitable scattering effects or were fully or partially reflective.

The applicant draws attention to the fact that the present invention may include any feature or combination of features disclosed herein either implicitly or explicitly or any generalisation thereof, without limitation to the scope of any of the present

claims. In view of the foregoing description it will be evident to a person skilled in the art that various modifications may be made within the scope of the invention.

CLAIMS

1. A light-emitting device comprising:

a pair of electrodes;

a light-emitting region located between the electrodes and comprising light-emitting organic material;

a first reflective layer and a second reflective layer, the second reflective layer being partially light-transmissive and the first and second reflective layers being located on either side of the light-emitting region to define a resonant cavity structure about the light-emitting region; and

a light-dispersal structure outside the cavity for dispersing light emitted from the cavity through the second reflective layer.

2. A light-emitting device as claimed in any preceding claim, wherein the light-dispersal structure comprises a matrix of light-transmissive material including a dispersion of localised regions of another material.

3. A light-emitting device as claimed in claim 1, wherein the light-dispersal structure comprises a grooved layer of light-transmissive material.

4. A light-emitting device as claimed in claim 1 or 2, wherein the light-dispersal structure comprises a ridged layer of light-transmissive material.

5. A light-emitting structure as claimed in any preceding claim, wherein the light-dispersal structure is capable of dispersing light emitted from the cavity substantially evenly over an angle of at least 90°.

6. A light-emitting device as claimed in any preceding claim, wherein at least one of the electrodes is light-transmissive.

7. A light-emitting device as claimed in any preceding claim, wherein at least one of the reflective layers is electrically conductive.

8. A light-emitting device as claimed in any preceding claim, having an encapsulant coating located between the second reflective layer and the light-dispersal structure.

9. A light-emitting device as claimed in any preceding claim, wherein one of the electrodes provides the first reflective layer.

10. A light-emitting device as claimed in any preceding clam, wherein the other of the electrodes provides the second reflective layer.

11. A light-emitting device as claimed in of claims 1 to 9, wherein at least one of the first and second reflective layers is formed as a distributed Bragg reflector.

12. A light-emitting device as claimed in any preceding claim, wherein the light-emitting organic material comprises a conjugated polymer.

13. A method for forming a light-emitting device having a pair of electrodes; a light-emitting region located between the electrodes and comprising light-emitting organic material; and a first reflective layer and a second reflective layer, the second reflective layer being partially light-transmissive and the first and second reflective layers being located on either side of the light-emitting region to define a resonant cavity structure about the light-emitting region; the method comprising forming a light-dispersal structure over the second reflective layer for dispersing light emitted from the cavity through the second reflective layer.

14. A method as claimed in claim 13, wherein the step of depositing the light-dispersal structure comprises forming a light-transmissive material including a dispersal of light-transmissive particles.

15. A light-emitting device substantially as herein described with reference to the accompanying drawings.

16. A method for forming a light-emitting device substantially as herein described with reference to figure 2 of the accompanying drawings.

ABSTRACT

LIGHT-EMITTING DEVICE WITH A LIGHT-DISPERSAL STRUCTURE

A light-emitting device comprising: a pair of electrodes; a light-emitting region located between the electrodes and comprising light-emitting organic material; a first reflective layer and a second reflective layer, the second reflective layer being partially light-transmissive and the first and second reflective layers being located on either side of the light-emitting region to define a resonant cavity structure about the light-emitting region; and a light-dispersal structure outside the cavity for dispersing light emitted from the cavity through the second reflective layer.

Figure 2

Fig. 1

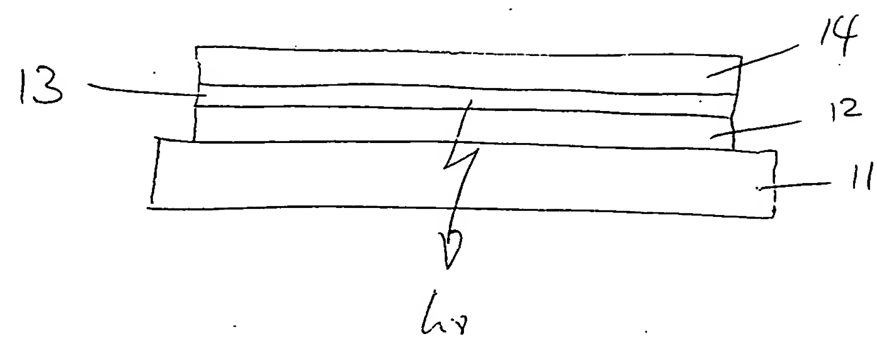

Fig. 2